



PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Toru NOGUCHI et al.

Group Art Unit: 1794

Application No.: 10/821,175

Examiner: E. COLE

Filed: April 9, 2004

Docket No.: 127794

For: CARBON FIBER COMPOSITE MATERIAL AND PROCESS FOR PRODUCING
THE SAME

DECLARATION UNDER 37 C.F.R. §1.132

I, TORU NOGUCHI, a citizen of Japan, hereby declare and state:

1. I completed doctoral courses in Materials Engineering at the Graduate School of Science and Technology of Kobe University in Hyogo, Japan in 1986 and obtained a BS in Materials Engineering which was conferred upon me by Tohoku University in Miyagi, Japan in 1977.
2. I have been employed by Nissin Kogyo Co., Ltd since 2002 where I have over 5 years of work and research experience in elastomers and metal composite materials. I was employed by Mitsuboshi Belting Ltd., from 1986 to 2001, where I had a total of 15 years of work and research experience, mainly in elastomer technology. I am a Guest Professor, Institute of Carbon Science & Technology, Shinshu University.
3. I am a member of The Japanese Society of Polymer Science, the Japan Institute of Metals and The Japan Society of Applied Physics.
4. I am a named inventor in the above-captioned patent application.
5. I have a professional relationship with the assignee of the above-identified patent application. In the course of that professional relationship, I received compensation

directly from the assignee for my work relating to Research and Development. I am being compensated for my work in connection with this Declaration.

6. I have extensive experience in, and knowledge of, the field of carbon fiber composite materials.

7. I conducted a series of tests to determine the properties of composites made from carbon nanofibers (CNT) with ethylene propylene diene rubber (EPDM).

8. The purpose of the tests were to examine the effect that the method of mixing the components had on the final composite product.

9. In these tests the same raw materials were used. Specifically, in the experiments EPDM 100 parts by weight (phr) was used as the primary elastomer. Carbon nanofibers with an average diameter of 13nm in the amount of 10 phr and peroxide (DCP) 2 phr were used as crosslinking agents.

10. Each identical set of materials was mixed using one of three different mixing methods (methods A-C).

11. Mixing method A was a shear rolling method using the machine shown in the attached Fig. 1 (attached). Method A is the mixing method described in Applicants' specification. Two open rolls in a machine, capable of being cooled or heated between 5-120°C, were used. The rotation speed of the respective rolls could range from 1-30rpm.

12. In the test conducted using Method A, the EPDM was inserted between the rolls and placed around the rolls. CNT was mixed with the EPDM into the gap of the rollers at room temperature for 10 minutes. The temperature was then increased to 100°C for a further 10 minutes of mixing. Finally, the mixture was subjected to tight milling five times at 20°C with a roll distance of 0.1mm.

13. Mixing method B was a general mixing method using a Labo Plastmill (Banbury mixer style). This mixer is shown in the attached Fig. 2. This machine has two

rotors and a chamber that can be heated. The chamber has a volume of 370 ml and a material filling fraction of 65%.

14. In the test conducted using Method B, the EPDM and CNT were inserted between the two rotors in the chamber. The front was closed and the materials were mixed at a rotor rotational speed of 30rpm. Three mixing cycles were conducted. The composite was taken out of the chamber at a temperature of 180°C.

15. Mixing method C used a twin screw extruder, shown in attached Fig. 3. The extruder had a screw diameter of 30mm and a cylinder diameter of 32mm. The length of the cylinder was 1344mm. The screws could reach a rotational speed of 524 rpm.

16. In the test conducted using Method C, the slots of the twin screw extruder were heated to a cylinder temperature of 100°C. The materials were placed between the cylinder and the screw and then mixed for 10 minutes. Once mixing was completed the composite was extruded from the twin screw extruder at a temperature of 180°C and cooled.

17. The results of the tests are shown in Table 1.

18. The column labels in Table 1 are as follows: T_B is the dynamic storage modulus; E_B is the elongation at break. T_{2n} and T_{2nn} are the spin-spin relaxation times, and are well defined in Applicants' specification. Finally, F_{nn} is the fraction of components having the second spin-spin relaxation time, and is also well defined in Applicants' specification.

19. The attached Figures collectively labeled as Fig. 4 are morphology observations of the sample prepared in the above described experiments. Figs. 4A1-4A3 show the results using Method A. Figs. 4B1 and 4B2 show the results of Method B. Finally, Fig. 4C show the results of Method C.

20. Figs. 4B1 and 4C show "sea island" structures, while Fig. 4A1 does not. These figures share a common white scale of 100 μ m. The sea island structures can easily be seen in Fig. 4B1 and Fig. 4C. The sea island structures are not present in Fig. 4A1.

21. These sea island structures are aggregations of carbon nanofibers, ranging in size from ten to hundreds of micrometers wide. The sea area is the EPDM matrix and the island areas are CNT aggregations.

22. Mixing methods B and C result in dispersion of carbon nanofiber aggregations. But mixing methods B and C do not result in uniform dispersion of the carbon nanofibers themselves. By contrast, Method A results in uniform dispersion of the carbon nanofibers.

23. Table 1 shows that Method A results in a first spin-spin relaxation time of 1860 μ s and a second spin-spin relaxation time of 6100 μ s. These values lie in the range defined in claim 1.

24. Table 1 shows that Method B results in a first spin-spin relaxation time of 3200 μ s and a second spin-spin relaxation time of 12000 μ s. These values lie outside the range defined in claim 1.

25. Table 1 shows that Method C results in a first spin-spin relaxation time of 3900 μ s and a second spin-spin relaxation time of 17000 μ s. These values lie outside the range defined in claim 1.

26. As indicated above, the same raw materials were used in each test. In each test the final product was a reinforced piece of composite material. Yet the spin-spin relaxation times of each composite material were substantially different.

27. I therefore conclude that the spin-spin relaxation times of a carbon nanofiber reinforced elastomer varies, at least, with the mixing method.

28. I also conclude that general mixing methods, such as the two comparative mixing methods used (Methods B & C) disperse CNT aggregations but do not disentangle CNT aggregates to uniformly disperse the carbon nanofibers.

I hereby declare that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine and/or imprisonment under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing therefrom.

Date:

May 7, 2009

Toru Noguchi
TORU NOGUCHI

Attachments:

Table 1

Figs. 1-4